Experimental determination of the L_2-L_3X Coster-Kronig transition probability in europium, plutonium, and curium*

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The L_2-L_3X Coster-Kronig transition probability for $Z = 63$, 94, and 96 has been measured by the L-x-ray—K-x-ray coincidence technique. The value measured in $_{63}$Eu was corrected for the presence of an unresolved $L\eta$ x-ray component in the $L\alpha$ x-ray group. The values reported are for $_{63}$Eu, $f_{23} = 0.172 \pm 0.015$, for $_{94}$Pu, $f_{23} = 0.233 \pm 0.015$, and for $_{96}$Cm, $f_{23} = 0.226 \pm 0.017$.

I. INTRODUCTION

Measurements of the L_2-L_3X Coster-Kronig transition probability, f_{23}, for elements $65 \leq Z \leq 96$ have been summarized in a recent review article.[1] These experimental values deviate significantly from theoretical estimates of Coster-Kronig transition probabilities.[2-5] Previous measurements exceed the calculated values by as much as 90%, with the average deviation being about 30%. For elements with $Z \geq 90$, this discrepancy may be explained by the fact that in this region, f_{23} is very sensitive to the details of the atomic wave functions since the L_2-$L_3M_{4,5}$ Coster-Kronig electron energies are very close to the threshold. However, the disagreement at other atomic numbers is not understood at this time.

The present measurements of f_{23} in $_{63}$Eu, $_{94}$Pu, and $_{96}$Cm were undertaken with high-resolution semiconductor detectors using the $L\alpha$-K coincidence technique. In the present measurement for $_{63}$Eu, the value of f_{23} was corrected for the presence of the unresolved $L\eta$ (L_2-M_1) line in the $L\alpha$ (L_3-$M_{4,5}$) x-ray group.

The reported value of f_{23} in $_{65}$Tb, $f_{23} = 0.066 \pm 0.014$,[6] is much lower than the predicted value, $f_{23} = 0.131$.[2] It was hoped that the present results for $Z = 63$ would determine if this were a significant trend in this region or perhaps some local anomaly at $Z = 65$.[7] Measurement of f_{23} at high Z is of particular interest since the jump in the value of f_{23} expected in the vicinity of $Z = 91$ gives insight into the Coster-Kronig electron energy.[8] The present measurements of f_{23} for Pu from the α decay of ^{243}Cm and Cm from the α decay of ^{249}Cf were undertaken with the hope of clarifying discrepancies between theoretical Coster-Kronig transition probabilities and the previously reported values at $Z = 94$ and 96.

II. EXPERIMENTAL PROCEDURES

The measurements of the L_2-L_3X Coster-Kronig transition probability at $Z = 63$, 94, and 96 were made using the K x-ray–L x-ray coincidence method described by Wood, Palms, and Rao[9] and others.[1] In this method a radioactive source which decays via internal conversion or electron capture (or both) provides atoms with vacancies in the K electronic shell creating K-L x-ray cascades. Vacancies in the L_3 subshell are indicated by the detection of $L\alpha$ x rays. In the K x-ray spectrum measured in coincidence with $L\alpha$ x rays, $K\alpha_1$ x rays indicate an L_3 vacancy which was created directly to fill the K shell vacancy and $K\alpha_2$ x rays indicate an L_2 vacancy which was created and followed by a Coster-Kronig transition which transferred the vacancy to the L_3 subshell. The ratio of coincidence counting rates of $K\alpha_2$ and $K\alpha_1$ x rays divided by the singles counting rates of these x rays yields the L_2-L_3X Coster-Kronig transition probability, f_{23}. The details of the calculation of f_{23} are described in Sec. III.

The Eu x rays were emitted from a 50-μCi source of 242-day ^{153}Gd which decays by electron capture to excited states of ^{153}Eu. The excited ^{153}Eu levels undergo K and L internal conversion. Decay by both electron capture and internal conversion leads to the emission of Eu x rays. The ^{153}Gd source was prepared by depositing $^{153}GdCl_3$ on 6×10^{-3}-mm-thick Mylar film.

The Pu x rays were obtained from a 100-μCi source of 32-yr ^{243}Cm which decays by α emission to several excited levels of ^{239}Pu. These levels deexcite via γ decay and internal conversion. The latter process leads to the emission of atomic x rays. Approximately 50% of the α decays of ^{243}Cm are followed by the emission of a K x ray.[10] The ^{243}Cm source was prepared by depositing a 1-mm-diam spot of $^{243}Cm(NO_3)_3$ on a 0.5-mm-thick aluminum disk covered by 2.5-mm beryllium. The Cm x rays were obtained from a 10-μCi source of 352-yr ^{249}Cf, 1 mm in diameter, deposited on a 0.15-mm aluminum disk and covered with 0.25 mm of beryllium. The x rays were emitted following the internal conversion of ^{245}Cm nuclear levels which were populated in the α decay of ^{249}Cf.

The K x rays were detected in a 10-mm planar Ge(Li) crystal with a sensitive depth of 5 mm. The crystal housing was fitted with a 0.12-mm-thick beryllium window. This detector, which had energy resolution of 500-eV full width at half-maximum (FWHM) at 122 keV, was able to completely resolve the $K\alpha_1$ and $K\alpha_2$ x rays of Pu and Cm. However, these same x rays in Eu were only partially resolved. Approximately 14% of the $K\alpha_1$ peak overlaps the $K\alpha_2$ peak of Eu. The $K\alpha$ peaks of Eu are shown in Fig. 1. The solid lines in this figure represent the resolved $K\alpha_1$ and $K\alpha_2$ peaks used in the data analysis. The peak shapes were determined by reflecting the low-energy half of the $K\alpha_2$ peak and the high-energy half of $K\alpha_1$ peak. Summation of the stripped peaks indicated an error of less than 2%. A total stripping error of 3% was used in the error analysis of the data.

The L x rays were detected in a 4-mm-diam planar Si(Li) crystal with a senstive depth of 3 mm. The crystal housing was fitted with a 0.05-mm-thick beryllium window. The energy resolution of the Si(Li) detector was 170 eV (FWHM) at 5.9 keV. K and L x-ray spectra of these isotopes detected with these crystals are given in Ref. 11. Both detectors were shielded laterally by tapered aluminum collimators, the use of which minimized the contribution of erroneous coincidences due to scattering of radiation from one detector to the other.

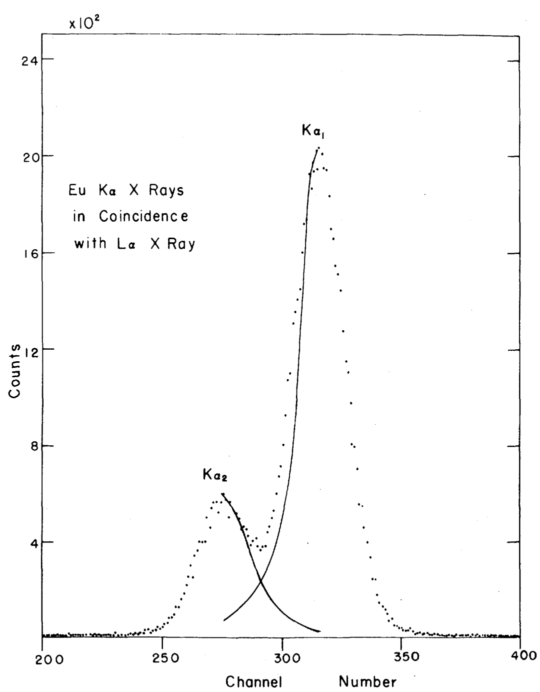

FIG. 1. Spectra of $K\alpha$ x rays of Eu in coincidence with $L\alpha$ and $L\eta$ x rays detected with the Ge(Li) spectrometer. The solid line indicates the resolved $K\alpha_1$ and $K\alpha_2$ peaks as explained in the text.

X-ray coincidence spectra were obtained with a standard fast-slow coincidence system employing crossover timing with a resolving time 2τ of 60 nsec. Pulses from the low-noise preamplifiers connected to each detector were sent to individual amplifiers with shaping time constants of 0.5 μsec for optimum timing characteristics. The bipolar output of each amplifier was sent to a separate timing single-channel analyzer (SCA). One SCA was set to pass all K x rays detected in the Ge(Li) crystal. The other SCA had discriminators set to pass the $L\alpha$ and $L\eta$ x rays in the Eu experiment but only the $L\alpha$ x rays in the Pu and Cm experiments. The preamplified signals from the Ge(Li) x-ray detector were sent to an additional amplifier with a shaping time constant of 2 μsec for optimum energy resolution. The K x-ray spectrum in coincidence with $L\alpha$ x rays was stored in a 1024-channel pulse-height analyzer. As a monitor of the experimental system the number of output pulses from each SCA and from the fast coincidence module were stored in separate scalers.

The K x-ray spectrum in coincidence with $L\alpha$ x rays was measured at an angle of 125° 15′ between the two detectors in order to avoid angular correlation effects.[11] Background determined in regions with no x-ray peaks was subtracted from each peak. The contribution from random coincidences was determined by taking measurements with an additional 640-nsec relative delay between the output of the two detectors and then removed. The extent of scattering of x rays and γ rays between detectors was determined by placing an absorber on the side of the source facing the L x-ray detector. In the Eu x-ray experiment the absorber was 0.50-mm aluminum, in the Pu and Cm x-ray experiments it was 0.50-mm copper. Contributions due to scattering were found to be negligible in all experiments.

III. DATA ANALYSIS

The value of f_{23} for ^{239}Pu and ^{245}Cm x rays was calculated as follows:

$$f_{23}=\frac{C_{K\alpha_2}(L\alpha)/S_{K\alpha_2}-C_{K\beta}(L\alpha)/S_{K\beta}}{C_{K\alpha_1}(L\alpha)/S_{K\alpha_1}-C_{K\beta}(L\alpha)/S_{K\beta}}, \qquad (1)$$

where $C_Z(Y)$ is the number of coincidence counts of x-ray Z in coincidence with x-ray Y, and S_Z is the number of singles counts of x-ray Z. The second term in both the numerator and denominator in Eq. (1), $C_{K\beta}(L\alpha)/S_{K\beta}$, is a correction term for the number of unwanted coincidence events due

TABLE I. A comparison between the energy separation of $L\alpha_1$ and $L\eta$ x rays and the resolution of the detector used in this work.[a]

Z	$E^{b}_{L\alpha_1}$	$E^{b}_{L\eta}$	ΔE^{c}	FWHM Si(Li) detector (eV)
63	5831	5848	17	175
94	14 290	16 460	2170	250
96	14 980	17 450	2470	265

[a]All energies in eV.
[b]Reference 12.
[c]$\Delta E \equiv E_{L\alpha_1} - E_{L\eta}$.

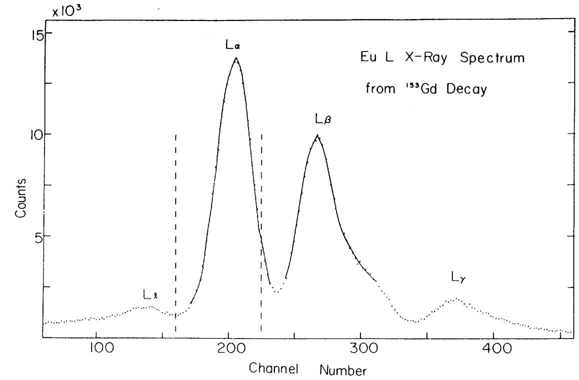

FIG. 2. Spectrum of L x rays of Eu detected with the Si(Li) spectrometer. The dashed lines indicate the position of the coincidence gate.

to a K and L x ray, each resulting from the migration of an electron hole formed in a different nuclear decay process in the same atom within a time shorter than the resolving time of the coincidence circuit. No correction for directional correlation effects is needed, as explained in Sec. II.

An additional correction to the ^{153}Eu data was made for the presence of $L\eta$ transition in the $L\alpha$ gate. The value of f_{23} for ^{153}Eu was calculated as follows:

$$f_{23} = \frac{C_{K\alpha_2}(L\alpha)/S_{K\alpha_2} - C_{K\beta}(L\alpha)/S_{K\beta}}{C_{K\alpha_1}(L\alpha)/S_{K\alpha_1} - C_{K\beta}(L\alpha)/S_{K\beta}} - \frac{[\Gamma(L\eta)/\Gamma_{L2}](\omega_2/\omega_3)}{\Gamma(L\alpha)/\Gamma_{L3}}, \quad (2)$$

where $\Gamma(X)$ is the radiative transition width for transition X, Γ_{Li} is the total radiative width of the L_i subshell, and ω_i is the fluorescence yield of the L_i subshell. The values of the radiative transition widths were taken from Scofield's calculations.[12] The value of the ratio ω_2/ω_3 used was interpolated from the theoretical calculations of Chen, Crasemann, and Kostroun[2] for neighboring elements and found to be unity. This calculation assumes that the $L\eta$ x ray is completely included in the $L\alpha$ gate. In Table I the energy difference between the $L\eta$ and $L\alpha_1$ x rays is compared to the energy resolution of the Si(Li) detector used in this study. At $Z = 63$, the $L\eta$ and $L\alpha_1$ x rays are unresolved while at $Z = 94$ and 96, the $L\eta$ x ray can easily be excluded from the $L\alpha$ x-ray gate.

TABLE II. Experimental data and theoretical predictions for f_{23} at $Z = 63$, 94, and 96.

Z	Predicted f_{23}	Reference	Parent isotope	Measured f_{23}	Reference
63	0.155	4			
	0.140	3			
	0.135	2			
			^{153}Gd	0.172± 0.015	present work
94	0.274	2			
	0.187	5[a]			
			^{242}Cm	0.22± 0.08	14
			^{242}Cm	0.21± 0.08	15
			^{242}Cm	0.229± 0.004[b]	18
			^{242}Cm	0.42± 0.08	16
			^{242}Cm	0.14± 0.08[b]	16 revised in 17
			^{243}Am-^{239}Np	0.226± 0.016[b]	7
			^{243}Cm	0.233± 0.015	present work
96	0.176	5			
			^{249}Cf	0.188± 0.010[b]	7
			^{249}Cf	0.226± 0.017	present work

[a]This calculation assumes L_2-$L_3M_{4,5}$ transitions are allowed.
[b]Error limits given are 2σ.

The ^{153}Eu data were also corrected for the presence of the tail of the $L\beta$ peak in the $L\alpha$ gate (see Fig. 2). This $L\beta$ contribution, found to be 0.3% of the total $L\alpha$ gate, had a negligible effect on the value of f_{23}. This contribution was considered in the error analysis.

IV. RESULTS AND DISCUSSION

The values of f_{23} for Pu and Cm calculated from Eq. (1) and for Eu calculated from Eq. (2) are given in Table II. All uncertainties are given as one standard deviation. Also given in Table II are the previously reported f_{23} values for Pu and Cm as well as theoretical predictions for f_{23} at $Z = 63$, 94, and 96. The value reported for f_{23} at $Z = 63$ is in reasonable agreement with a prediction based on a linear interpolation between the calculated values for f_{23} at $Z = 60$ and 67 by Chen and Crasemann.[4] However, the measured value is significantly higher than the earlier predictions by both Chen, Crasemann and Kostroun[2] and a value interpolated from McGuire.[3] The present measurement at $Z = 63$ suggests that the reported value[6] at $Z = 65$ is much too low. This conclusion is also supported by the recent measurement at $Z = 65$ by Rao and coworkers.[13] The present value for f_{23} at $Z = 94$ agrees well with the values previously reported for f_{23} for different radioactive sources.[8,14-18] However, both the present value and the value obtained by McGeorge *et al.*[8] and Campbell *et al.*[18] are significantly higher than the prediction of McGuire[5] and lower than the value predicted by Chen, Crasemann, and Kostroun.[2] The present value at $Z = 96$ is significantly higher than both the previously reported value[8] and the theoretical prediction by McGuire. In conclusion, the present values for f_{23} at $Z = 63$, 94, and 96 are higher than most theoretical predictions, which is similar to the discrepancy between the measured and calculated values of this Coster-Kronig transition probability in other elements.

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